REFRACTORIES FOR THE GLASS INDUSTRY

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EFFECT OF THE GLASS COMPOSITION ON CORROSION OF ZIRCONIUM-CONTAINING REFRACTORIES IN A GLASS-MELTING FURNACE (A REVIEW)

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Different chemical compositions of glasses and the individual characteristics of their reaction with zirconium-containing refractories are examined. When such refractories are used for lining glass-melting furnaces, it is necessary to consider the chemical compositions of the glass which will be melted in these furnaces.

The following factors that act simultaneously and together affect corrosion of refractories, including zirconium-containing refractories, in the glass-melting furnace:

quality of the refractory (chemical and mineral compositions, structure, properties);

design of the glass-melting furnace;

characteristics of the heat insulation;

output of the furnace;

arrangement of the refractories in the furnace;

type and distribution of fuel over the volume of the furnace;

temperature-time conditions of melting;

intensity of exchange of the glass melt in the furnace (dependence on outputs, convective flows, presence of supplementary electric heating, bubbling, etc.);

stability of manufacturing process;

redox conditions of melting;

type of raw materials and impurities in them;

redox potential (ORP) of raw materials;

characteristics of new types of raw material;

batch composition and ORP;

batch : cullet ratio

concentration and type of auxiliary raw materials;

volatility of the components of the glass;

chemical composition, acid-base character, and ORP of the glass.

All of these factors are examined to some degree in [1 – 10].

The chemical composition, acid-basic character, and ORP of the glass are especially important in corrosion of refractories, including zirconium-containing refractories.

We do not claim completeness in discussing the problem here due to the large number of studies in this area and the ambiguity of interpreting the results, but we believe it useful to direct the attention of process engineers in the refractories field and glass process engineers to the complexity of this problem, which is a function of the quality of the refractories, the variety of the glass compositions, their tendency to constantly vary, including the initial raw materials.

If we consider the glass compositions, there is almost no element in the periodic table which has not been "dropped in" the composition of glass. Rare-earth, heavy elements, halogens, and even gaseous components (N, H, S, F, and others) incorporated in the structure of the glass can be found in glass compositions. For this reason, the variety of glass compositions is much larger than the assortment of refractory materials suitable for quality glass melting, although most industrial glass production is for sheet glass and bottle glass (OST 21-51–82) which have similar chemical compositions (Table 1).

Zirconium refractories belong to the acid class, since SiO_2 and ZrO_2 have acid properties, while Al_2O_3 has amphoteric properties. For this reason, the zirconium refractory is most stable in alkali-free and low-alkali glasses, i.e.,

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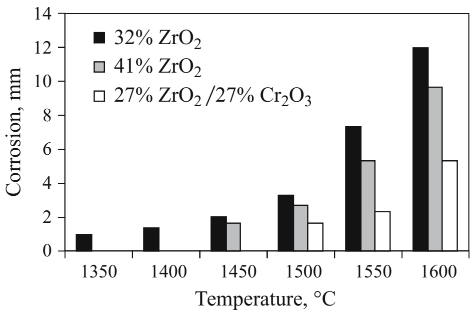

Fig. 1. Corrosion of electric smelting refractories as a function of temperature [5].

with a low basicity ratio, which in a first approximation can be estimated with the equation:

$$M_{bas} = \frac{\sum Me_nO_m}{R_nO_m},$$

where ΣMe_nO_m and ΣR_nO_m are the total content of basic and acid oxides, respectively.

More complex equations for calculating the basicity ratio can be found in [11], for example.

However, higher temperatures are necessary to melt glass with a low basicity ratio, and this also negatively affects the glass stability of a refractory.

In addition to the acid-base properties of a glass melt, the anion structure coefficient, which takes into account the number of O^{2-} ions with the highest activity, and the amount of glass-forming components that complicate the structure of the melt are to evaluate the corrosiveness [12]. If the value of this coefficient is low (less than 2.85), the corrosiveness is also relatively low; at higher values (especially greater than 3), the resistance of the refractory decreases sharply.

The glass resistance of zirconium-containing refractories has been repeatedly investigated by many researchers [1, 13, 14]. For example, the effect of the temperature on the corrosion rate is shown in Figs. 1 and 2.

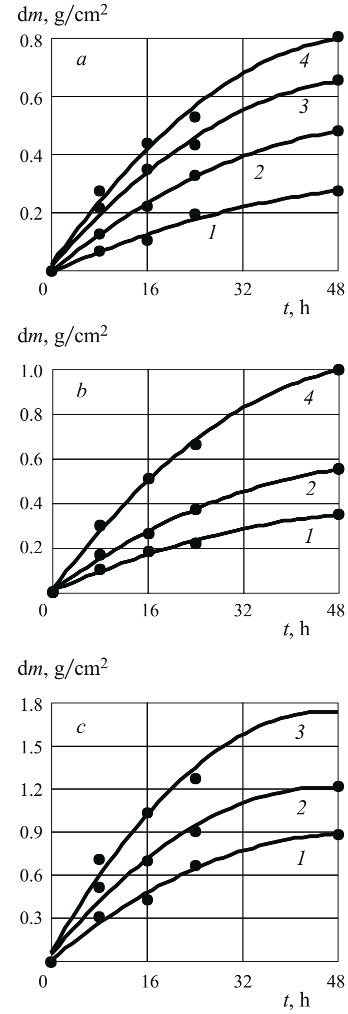

Fig. 2. Amount of dissolved refractory dm in sodium-calcium-silicate glass melt as a function of time t in dynamic tests [1]: *a* and *b*) Bakor-33 at 1400 and 1550°C; *c*) Bakor-41 at 1550°C; *1*, *2*, *3*, and *4*) sample rotation rate in melt of 20, 40, 80, and 120 min^{-1}.

In industrial conditions, it was found on the float-glass line that increasing the temperature 50°C (from 1550 to 1600°C) at specific outputs of 1500 – 2000 kg/m^2 a day re-

TABLE 1

Glass group	Mass content, %						
	SiO_2	Al_2O_3	CaO	MgO	$Na_2O + K_2O$ or Na_2O	Fe_2O_3, max	SO_3, max
Float glass	72.4	1.4	9.2	3.4	13.3	0.15	0.4
Bottle glass:							
colorless BT-1	72.0	2.4	11.0		14.0	0.1	0.5
off-white PT-1	71.4	2.5	11.0		14.2	0.5	0.4
green ZT-1*	70.3	2.2	11.0		14.3	0.8	0.3
brown KT-1	71.1	2.8	11.0		14.3	0.5	0.3

* Including 0.1% Cr_2O_3.

duces the time for attaining the residual thickness of the refractory in a tank furnace (30 mm) by 30 – 35%, while increasing output to 500 kg/m^2 a day (from 1500 to 2000) at a melting temperature of 1550 – 1600°C only reduces the time by 7.9% [15].

Corrosion of a refractory in the aggressive multicomponent glass melt (sheet, bottle) consists of formation of reaction zones with a subsequent increase in the amount of melt in the volume of the refractory, which finally causes its destruction [12]. The kinetics of the reaction is basically determined by the counter diffusion of the components of the refractory and melt [16].

Na_2O is most intensively incorporated in a refractory while Mg, Ca, and Si oxides have lower mobility. The following order of the mobility of the components was observed (components inert in the intermediate layer are indicated in parentheses) [16]: Na_2O, MgO, CaO, SiO_2 (Al_2O_3, ZrO_2, Cr_2O_3).

The dependence of the corrosion rate K on the temperature is exponential, i.e., it obeys the Arrhenius equation [1, 17]:

$$K = K_0 e^{\frac{-E}{RT}},$$

where K_0 is a preexponential factor; R is a constant; T is the thermodynamic temperature; E is the activation energy (calculated value).

The higher the corrosive stability of a refractory, the higher the activation energy [17]. According to tests at 1400 – 1550°C for 120 h in static conditions, the following values were obtained.

Calculated Activation Energy of Corrosion for Different Glass Compositions [17]

Glass of the system	Activation energy, kJ/mole
$Na_2O - CaO - SiO_2$	344 – 427
$BaO - SiO_2$	247 – 344
$PbO - SiO_2$	344 – 390
$B_2O_3 - SiO_2$	~ 250
$CaO - Al_2O_3 - SiO_2$	201 – 293

The activation energy ranges from 200 to 400 kJ/mole for the different glass compositions, and this corresponds to a change in corrosion by 1.5 – 2 times when the temperature changes by 50°C in the 1300 – 1600°C range [7, 18].

These data show that B_2O_3, followed by Na_2O, CaO, PbO, and BaO, are the most aggressive components with respect to the zirconium-containing refractory. For example, corrosion of Bakor-33 in borosilicate glass was 110 mm after 2 months of operation of the glass-melting furnace, i.e., 0.6 mm a day, and the furnace runs in melting such glasses are 4 – 8 months [19]. The glass compositions of the $Na_2O - CaO - SiO_2$ system can be assigned to the **first**

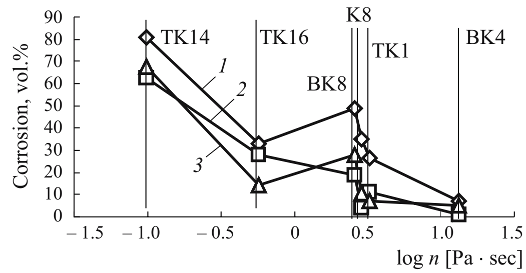

Fig. 3. Corrosion of some refractories as a function of melt viscosity [22]: TK, BK, K) grades of optical glass; *1*) mullite–corundum; *2*) BKCh-33; *3*) Kor-95.

group of compositions; they are used for large-tonnage production of sheet and bottle glass.

The following tendencies (immediate) are observed when sheet and bottle glass compositions and technology are changed:

a decrease in the maximum melting temperature of large-tonnage glass due to an increase in the mass content of alkaline components to 17%; since this will be accompanied by a decrease in the chemical stability of the glass, small amounts of B_2O_3 and K_2O, which create a “polyalkaline” effect with Na_2O, can be added;

conversion to dolomite-free and magnesium-free compositions with an increase in the CaO content in the glass compositions.

In both cases, the aggressiveness of the glass melt increases, although this is probably compensated by the lower melting temperature.

The **second large group** of glass compositions for which zirconium refractories are used in melting consists of borosilicate and other glasses with elevated chemical and thermal stability. Such glasses are used for fabrication of medical [20], chemical-laboratory [21], optical, and other articles. The chemical compositions of some borosilicate glasses [20, 22] are reported in Table 2.

Borosilicate melts are distinguished by high aggressiveness to zirconium-containing refractories, including types ER 1711 [1, 19, 23]. In this case, corundum refractory materials are more resistant (Fig. 3) [20, 22]. For example, in melting borosilicate photochromic glass in an electric furnace in which the interior temperature is 1350°C and the temperature on the surface covered with the batch is 1200°C, the maximum degree of corrosion in the upper part of the tank reached 110 mm after 2 months of operation, and the average corrosion rate was 1.8 mm/day [19].

The duration of the run of gas-flame glass-melting furnaces with output of 30 tons/day with Bakor refractories varies from 6 months to 3 years [20].

Some believe that some borosilicate melts and phosphate melts are more resistant to zirconium-containing refractories if the mass content of Na_2O does not exceed 11% [24, 25]. For this reason, zirconium refractory of high purity is of great interest in melting many special, including optical,

TABLE 2

Glass group	Mass content, %								
	SiO_2	Al_2O_3	B_2O_3	CaO	MgO	Na_2O	K_2O	BaO	other
Medical glass:									
NS (1, 2, 2A, 3)	72.8 – 73.0	3.5 – 4.5	2.5 – 6.0	5.0 – 7.0	0.8 – 3.0	8.5 – 11.0	1.7 – 2.0	–	–
KhT	74.0	5.0	8.0	1.0	0.2	5.0	2.8	4.0	–
Type E glass fiber	53.0	15.0	10.0	17.0	4.0	0.3	–	–	–
Photochromic glass	60.1	9.5	20.0	–	–	10.0	–	–	–
Primer enamel	58.0	4.0	19.3	4.0	–	14.0	–	–	0.7
Cover-coat enamel	48.2	–	13.2	–	–	7.4	1.4	–	12.6 Na_2SiF_6 13.2 CeO_2
Chemical-laboratory KS [33]	67.0	2.15	3.9	6.3	2.27	12.6	2.65	–	0.87 ZrO_2 2.26 ZnO
Some optical glasses:									
K8	73.8	–	9.8	–	–	15.0	1.3	0.1	
BK4	74.3	–	3.3	–	–	11.0	7.0	4.4	
BK8	64.5	–	15.5	–	–	7.1	9.5	3.4	
TK1	63.6	–	11.8	–	–	4.5	15.6	4.5	
TK14	47.9	–	21.7	–	–	0.7	25.8	3.9	
TK16	52.4	–	12.2	–	–	–	28.2	7.2	

glasses [26]. However, the alkaline-earth oxides in any glass composition destroy a refractory more strongly than alkaline oxides [13, 22]. For this reason, in high-calcium glass compositions (20 wt.% CaO and higher), zirconium-containing refractories are insufficiently stable. We should also note that melts with a high B_2O_3 content destroys refractory B-90 more intensively than refractory ER 1711. At the same time, a glass melt containing the same amount of alkali metal oxides, but where B_2O_3 is replaced by Al_2O_3, on the contrary intensively destroys refractory ER 1711 and not high-zirconium B-90 [13].

The **third group** of compositions used in mass production are opal and milk glasses for illumination engineering, high-quality, and other kinds of articles that contain up to 70 wt.% fluorine, which, like other halogens, increases the aggressiveness of the melt toward the refractory. The compositions of some fluorine-containing industrial glasses [27] are reported in Table 3.

The furnace runs for melting fluxes whose melts contain up to 6 wt.% fluorine compounds that intensively erode the tank lining are a maximum of 2 – 3 months [26].

Lead-containing glasses are a special glass group not only with respect to the chemical composition but also the aggressiveness toward refractories and the toxicity. This group includes high-quality glass (crystal) [21] and many optical glasses and glasses that protect against short-wave radiation [18, 28, 29]. The compositions of some lead-containing glasses [18] are reported in Table 4.

The dependence of the activity of the glass melt on the lead oxide content is shown in Fig. 4.

Corrosion of a zirconium-containing refractory in lead-aluminosilicate glass with a 36% mass content of PbO is 2.5 times higher than in glass containing 14% PbO. When the

TABLE 3

Glass	Mass content, %						
	SiO_2	Al_2O_3	B_2O_3	CaO	K_2O	Na_2O	F
Milk:							
opacified with fluorine	64.3	9.2	–	2.7	1.0	16.3	6.5
opacified by liquation	73.0	1.6	9.2	11.1	–	5.1	–
Opal*	59.3	12.7	–	1.0	3.0	11.0	3.0
Cover-coat enamel**	44.3	4.6	6.8	–	2.9	12.3	17.4 Na_2SiF_6 2.8 CaF_2

* Including 10.0% PbO.
** Including 4.1% ZnO.

TABLE 4

Glass group	Molar content, %			
	SiO_2	ÐbÎ	K_2O	B_2O_3
Optical:				
VS92	72.0	14.0	10.0	–
TF5	61.0	36.0	–	–
STF11	20.0	65.0	–	12.0
High-quality (crystal)	57.5	24.0	15.5	1.0

PbO content increases to 65% (glass with x-ray and γ-radiation protection), the corrosion increases by 15 – 30 times, although such glasses melt at a low temperature — around 1000°C [18].

Many optical glasses do not have a silicate, but instead a phosphate base as the glass former. For example, the composition of phosphate crown is as follows (mass content, %): 3.0 B_2O_3, 10.0 Al_2O_3, 12.0 K_2O, 4.0 MgO, 0.5 As_2O_3, 70.0 P_2O_5. Phosphate glasses are more acid than silicate glasses. Erosion of refractories is less in phosphate glass than in multilead and lead-silicate glass [25]. In addition to the indicated components, optical glasses contain lanthanum, indium, tungsten, antimony, neodymium, and other oxides.

Note the content of iron oxides in the glass compositions (up to 0.15 wt.% in sheet glass) that can be present as an undesirable impurity from the raw materials and intensify corrosion of refractories [12, 30]. However, iron oxide in larger amounts acts as a dye in production of heat-shielding glass (approximately 1 wt.%) or an equivalent component when its content is up to 10% and higher. In all cases, it is important to shift Fe(II) ↔ Fe(III) to the required side, which is very important in manufacturing processes and in production of high-quality glass (spectral characteristics, absence of defects).

Iron oxides are now used not only in bulk-dyed heat-shielding glass technology. Basalt fiber, metallurgical slags in bottle-glass technology, iron-containing glass crystal materials, and semiconductors are a far from complete list of iron-containing glasses [12]. In glass-melting practice, great attention has recently been focused on studying the ORP of the raw material, batch, and the redox conditions of the melt which affect shifting of the Fe(II) ↔ Fe(III) equilibrium. A glass melt that contains Fe(II) is more aggressive toward refractories than a glass melt with Fe(III) [30, 31].

The dependence of the corrosion of a refractory in an iron-containing melt on the temperature is shown in Fig. 5. It is necessary to consider that the temperature, like intensification of the acid properties of glass, shifts the equilibrium toward Fe(II) [30].

Oxygen blowing is now used for glass melting in industrialized countries to enhance glass melting and reduce atmospheric emissions of nitrogen oxides. This naturally alters the redox conditions of glass melting and can shift the equilibrium toward the less aggressive Fe(III).

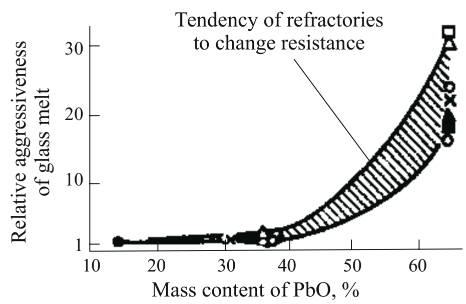

Fig. 4. Effect of PBO content in glass on corrosion of refractories [18]: □) BKCh-33; ■) KÉL-95; ×) leucosapphire; ●, ○, ▲, △) aluminosilicates.

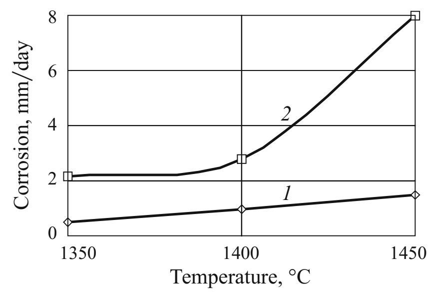

Fig. 5. Effect of the temperature on the corrosion rate of refractories in a melt containing (wt.%): 50 SiO_2, 18 Al_2O_3, 22 CaO, 10 MgO; in excess of 100%: 0.3 Cr_2O_3, 2.5 Na_2O, 5.0 – 15.0 Fe_2O_3 [31]; *1*) KhATs-30; *2*) BK-33.

These glass compositions are manufactured in relatively large volumes. The compositions of special types of industrial glass were not examined, since zirconium-containing refractories are basically not used in their production.

Despite the variety of glass compositions, there is one general rule: corrosion of refractories increases with a decrease in the viscosity of the glass melt. This is also seen in optical glasses (see Fig. 3) [22]. An increase in the the melting temperature is always accompanied by a decrease in the viscosity of the glass melt.

In evaluating corrosion of refractories, it is necessary to consider how the components of the glass affect the viscosity. Of the traditional components in mass-produced glass compositions, alkaline components decrease the viscosity to the greatest degree in the following order: $Li^+ \rightarrow Na^+ \rightarrow K^+$, followed by alkaline-earth components. Glass-forming oxides decrease the viscosity in the order: quartz → silicate (germinate) → borate (phosphate) → fluoride → chalcogenide glass. For example, B_2O_3 and PbO decrease the viscosity of glasses of the system $Na_2O – CaO – B_2O_3 – PbO – SiO_2$. However, if the same glass composition contains Al_2O_3 instead of B_2O_3, refractory ER 1711 is more intensively affected than the high-zirconium refractory [13].

TABLE 5

Glass types	Recommended clarifiers and combinations	Recommended clarifier content, wt.%	Incongruous combinations of clarifiers
Soda-lime (sheet, bottle, high-quality)	Na_2SO_4, $BaSO_4$, $(NH_4)_2SO_4$, NaCl, NH_4Cl	0.5 – 3.0	$Na_2SO_4 + Na_2SiF_6$
Industrial and high-quality	$As_2O_3 + KNO_3$, $Ba(NO_3)_2$, $Sb_2O_3 + KNO_3$, $CeO_2 + NaCl + Ba(NO_3)_2$, $Ca(NO_3)_2$, $Na_2SO_4 + NaNO_3$, $Na_2SO_4 + CaF_2 + NaCl$, $BaSO_4 + CaF_2 + NaCl$	1.0 – 3.0 NaCl, 1.0 – 4.0 CaF_2, 0.5 – 1.0 Na_2SO_4, 0.1 – 0.5 As_2O_3, 3.0 – 5.0 $Ba(NO_3)_2$, 1.0 – 3.0 KNO_3, 0.5 CeO_2, 0.1 – 1.0 Sb_2O_3	$Na_2SO_4 + As_2O_3$
Crystal and lead-containing glass	$As_2O_3 + KNO_3$	1.0 – 3.0	–
	$Ba(NO_3)_2$, $NaNO_3$	0.1 – 0.5	
	NH_4NO_3	1.5 – 3.5	

It is also necessary to consider that the effect of these glass compositions on refractories was examined with respect to the main components. Auxiliary components play an important, and sometimes also a fundamental role in the reaction of the glass melt with the refractory. They include clarifiers, glass melting accelerators, dyes, and oxidizing and reducing agents [6, 27, 32]. Glass-melt clarifiers and their optimum concentrations [33] are reported in Table 5.

Corrosion of refractories is thus complexly dependent on the composition of the glass melt: on the type of component in the glass, its quantitative content, the overall composition of the glass, and the ratio of the components. It is necessary to consider all parameters of glass-melting technology.

There are now many studies on the complex use of the feedstock and replacement of some traditional raw materials by secondary products from different industrial sectors. These materials are most frequently not only unstable in chemical composition but can also contain impurities that significantly intensify the aggressiveness of the glass melt with respect to the refractory.

In addition to the listed materials and other combinations of auxiliary materials which can activate the reaction of the glass melt with the refractory are also used for other glass compositions and glass crystal materials. Small impurities in the raw materials must also be taken into consideration, especially in secondary products. For example, in the 1980s – 1990s, in view of the shortage and high price of soda, attempts were made to find substitutes for it from a series of secondary products. Soda melt — the product of combustion of caprolactam production waste — was interesting. Its chemical composition (mass content, %) is: 95.0 – 98.0 Na_2CO_3, 0.6 – 0.7 Na_2SO_4, 0.5 – 0.8 NaOH, traces of NaCl, Cr_2O_3, CoO [33, 34]. Use of soda melt instead of bottle glass standard soda in two glass works without preliminary studies led to premature shut-down of the glass-melting furnaces for maintenance due to erosion of the refractories up to breakthrough of the glass melt.

We studied the glass resistance of the refractory to a glass melt from a batch with soda melt and found that replacement of standard soda by 40% soda melt does not affect the glass resistance of the refractory. On this basis, soda melt in the corresponding ratio with standard soda (40 : 60) was introduced under our surveillance in an industrial plant and had a significant economic effect [33, 34].

The ideal version is thus 100% glass resistance of refractories. However, this is not the case at present and will scarcely be the case anytime in the future.

The most intensive corrosion of the refractory is observed in the tank of the glass-melting furnace at the site of contact with the batch and surface layer of the glass melt, although according to the Rules for Industrial Operation of Plants [35, 36], fluctuations in the level of the glass melt, for example, in continuously operating, large-tonnage furnaces, should not exceed ± 0.2 – ± 0.5 mm.

It was recently found that with respect to corrosion of refractories, not only the tank, but also other elements of the lining (feed hopper, burner openings, arches, barriers). For this reason, we recommend that more glass-resistant refractories be installed in individual parts of the furnaces in comparison to electrosmelting zirconium-containing refractories.

For example, more glass-resistant chromium–aluminum–zirconium refractories are recommended for the top level of the glass-melting furnace in the melting and maximum temperature zone and in the region of the first pair of burners and flow supply beams. However, they can only be used for a colored glass melt, for example, for green and brown bottles, since chromium impurities in the amount of less than 1 wt.% intensively color the glass and have limited solubility in silicate melts [19].

The job of specialists in the refractories sector for glass-making is to increase their glass resistance by developing the optimum compositions and production technology to improve the properties. It should be taken into consideration that specialists in glass melting will attempt to develop better designs for glass-melting furnaces, improve the compositions and production technology for existing types of glass, and develop new types as a function of the needs of science and technology and in consideration of energy and resource conservation.

To increase the duration of the furnace runs, i.e., to prolong the refractory servicing time, air cooling of the tank walls [2, 3, 19, 37], hot repairs [38], ceramic fusing

(Stekloinvest Co., Stavropol' Region; FOSBEL, Germany, etc.), and evaporative cooling of the furnaces are used [8, 19, 26, 39 – 41].

Evaporative cooling of the tank walls allows extending the servicing time by 2.5 – 3 times even in melting an aggressive glass melt [26]. However, use of evaporative cooling slightly increases fuel consumption.

The major possibility of organizing heat shielding of the refractory lining of the walls of the melting tank from contact with the batch [9, 10] was demonstrated to ensure melting in the near-wall zone with additional fuel feed (short-flame burners or supplementary electric heating — SEH) in the amount of 2.0 – 2.5% of the total consumption for a furnace with output of approximately 300 tons of glass melt a day [10]. However, the use of SEH in melting aggressive glasses in a tank laid out on Bakor beams can in many cases intensify the corrosion [13].

In assessing the glass resistance of refractories in industrial conditions, especially in emergency situations, and in improving the technology for production of both refractories and glass, knowledge of each technology and mutual understanding among specialists in the glass melting and refractory technology sectors are necessary.

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